

Femtosecond pulse damage thresholds of dielectric coatings in vacuum

Duy N. Nguyen,¹ Luke A. Emmert,¹ Paul Schwoebel,¹ Dinesh Patel,² Carmen S. Menoni,² Michelle Shinn,³ and Wolfgang Rudolph^{1,*}

¹Department of Physics and Astronomy, University of New Mexico, Albuquerque, NM 87131, USA

²Department of Electrical and Computer Engineering, Colorado State University, Fort Collins, CO 80523, USA

³Free Electron Laser Division, Jefferson National Accelerator Lab, Newport News, VA 23606, USA
[*wrudolph@unm.edu](mailto:wrudolph@unm.edu)

Abstract: At 10^{-7} Torr, the multiple femtosecond pulse damage threshold, $F(\infty)$, is about 10% of the single pulse damage fluence $F(1)$ for hafnia and silica films compared to about 65% and 50%, respectively, at 630 Torr. In contrast, the single-pulse damage threshold is pressure independent. The decrease of $F(\infty)$ with decreasing air pressure correlates with the water vapor and oxygen content of the ambient gas with the former having the greater effect. The decrease in $F(\infty)$ is likely associated with an accumulation of defects derived from oxygen deficiency, for example vacancies. From atmospheric air pressure to pressures of $\sim 3 \times 10^{-6}$ Torr, the damage “crater” starts deterministically at the center of the beam and grows in diameter as the fluence increases. At pressure below 3×10^{-6} Torr, damage is initiated at random “sites” within the exposed area in hafnia films, while the damage morphology remains deterministic in silica films. A possible explanation is that absorbing centers are created at predisposed sample sites in hafnia, for example at boundaries between crystallites, or crystalline and amorphous phases.

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OCIS codes: (140.7090) ultrafast lasers, (140.3330) laser damage, (310.6870) thin films, other properties.

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1. Introduction

An increasing number of high-power laser applications involve optical components with dielectric coatings that are operated under vacuum conditions. Examples are space-based lasers [1], high-power femtosecond pulse lasers [2], and interferometers such as the Laser Interferometer Gravitational-Wave Observatory (LIGO) [3,4]. Previous studies with nanosecond lasers under vacuum and sealed atmospheric conditions have shown that the useful lifetime of the optics and multiple pulse damage fluence reduced significantly for dielectric films [5-8] and bulk materials [9,10] compared to atmospheric conditions. Two explanations have been given for these observations. The first involves the deposition of absorbing graphite layers from photolytic cracking of background organic contaminants that result from out-gassing of epoxies [7]. These deposits are reduced by the presence of oxygen in the atmosphere, but grow unchecked in sealed systems or under vacuum. The other observation of reduced damage resistance was associated with the absorption of water vapor by films [8]. It was hypothesized that local strain in the film induced by adsorbed water reduces their damage threshold. In both cases, the effects of ambient gas on the laser damage threshold are more prominent in films prepared by electron-beam evaporation. When using high-density films such as those prepared by ion-assisted deposition, the deposition rate of graphite is reduced by a factor of 50 [5] and the effect of water eliminated [8].

These previous studies of the effect of vacuum on damage resistance were performed with nanosecond pulses and repetition rates of 10-100 Hz. The effect of ambient pressure on

damage by femtosecond (fs) pulses has not been reported. The fs pulse damage behavior under atmospheric conditions has been well explained [11,12]. The damage processes are highly deterministic and result from dielectric breakdown at the location of highest laser intensity [13]. Explanation of multiple pulse damage phenomena requires the consideration of laser-induced material modifications by sub-threshold pulses, but the results are similarly deterministic [12].

We report here on the fs pulse dielectric breakdown behavior of optical coatings under low pressure conditions for various ambient gases. The studies were performed on ion-beam sputtered hafnia and silica films, which form the backbone of optical interference coatings. The drop in damage threshold is dependent on water vapor and oxygen pressures, but for reasons different than discussed previously for nanosecond pulses. In hafnia films, we also observed a change in the damage mechanism from a deterministic process at high pressure to a stochastic process for pressures below a few 10^{-6} Torr. Our results are of particular interest for high-intensity femtosecond laser interactions with coated optical components under low pressure (< 1 Torr) conditions as are typical for terawatt and petawatt systems [2].

2. Experimental

The threshold fluence for dielectric breakdown (damage) was measured for various gas pressures and atmospheres. A diagram of the experimental setup is shown in Fig.1(a). A fs Ti:sapphire oscillator-amplifier system produced 800-nm, 1-kHz pulse trains containing an adjustable number of pulses. The pulse duration in all experiments was set by the compressor stage of the amplifier to 50 fs. The energy of the pulses was controlled coarsely with an attenuator inside the amplifier, while fine tuning was done by a pair of in-line counter-rotating, glass plates near Brewster's angle. The energy of each individual excitation pulse was measured with a calibrated photodiode. The samples were placed inside a vacuum chamber and the pulses were focused through a fused silica window. The waist of the Gaussian beam, w_0 at the sample surface was about 20 μm .

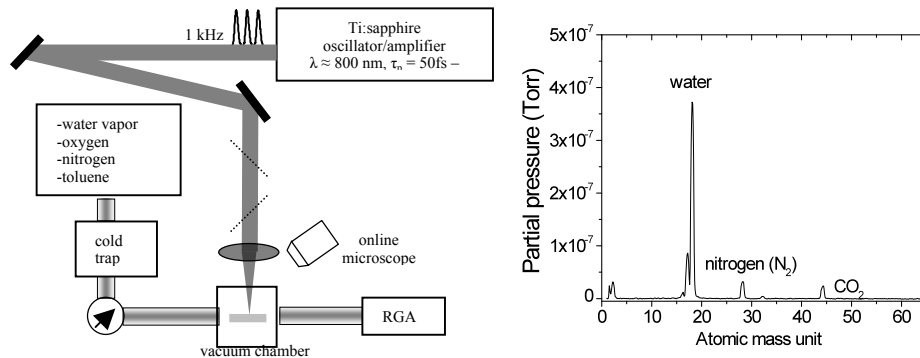


Fig. 1. (a) Schematic diagram of the fs dielectric breakdown (damage) measurements in various ambient gas environments and at different pressures. (b) RGA spectrum at a base pressure of 3×10^{-7} Torr.

The chamber was evacuated by a turbo-molecular pump to a base pressure of 3×10^{-7} Torr. The pressure and gas composition for the experiment was set by introducing different gasses of interest through a variable-rate leak valve. A cold trap removed residual water vapor in the gas line when necessary. A Residual Gas Analyzer (RGA100, Stanford Research Systems) allowed us to analyze the partial pressure of gaseous components inside the chamber as long as the total pressure was below 10^{-4} Torr. Figure 1(b) shows a typical RGA spectrum at a base pressure of 3×10^{-7} Torr, indicating that the chamber was essentially free of organic contamination with partial pressures above 10^{-8} Torr.

Film damage was observed with a microscope and CCD camera (online) by monitoring the change of scattered light at the illuminated spot. A round-robin experiment comparing

different damage diagnostic techniques showed that this method is accurate within acceptable error bars [14]. The damage morphology of the films was studied *ex situ* with a Nomarski microscope (Olympus BX60).

For the single-pulse experiments (1-on-1) the sample was moved after each pulse regardless of whether visible damage occurred to avoid any pulse accumulation effects. For multiple pulse experiments (S-on-1), the sample was illuminated with a burst of a pre-selected number (S) of pulses. To obtain the multiple-pulse threshold defined here as $F(\infty) = F(S=\infty)$ the pulse train continuously illuminated the sample. $F(\infty)$ was determined as the smallest fluence at which no damage occurred after 5 minutes (300,000 pulses) of illumination. Pulse-to-pulse energy fluctuations of the laser system were approximately 2%.

Unless stated otherwise the samples were quarter-wave ($\lambda = 800$ nm) thick hafnia (HfO_2) films deposited on super-polished fused silica substrates using dual ion-beam sputtering (DIBS) with a hafnium-metal target [15].

3. Results

Figure 2 shows the breakdown fluence as a function of the number of pulses illuminating one and the same sample site at atmospheric pressure (630 Torr at an elevation of 1,600 m) and for 3×10^{-7} Torr. The data at atmospheric pressure show the expected behavior [16]. The damage fluence drops with increasing number of pulses until it levels off at a value $F(\infty)$, which is a few tens of percent below the value for single pulses. This drop can be explained with the occupation of native and laser induced defects during the pulse train and their re-excitation by subsequent pulses [11,12,16]. Values of $F(\infty)$ for transparent materials are commonly 50-80% of the single-pulse value [11,12,17]. At 3×10^{-7} Torr, the measured values of $F(S)$ are the same as for 630 Torr for $S < 300$, but drop off dramatically for larger number of pulses. $F(\infty)$ reaches a value of 10% of the single-pulse value, which indicates the presence of additional processes that affect the damage at low pressure.

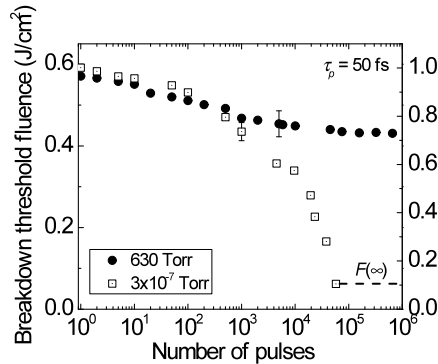


Fig. 2. Damage fluence as a function of pulse number exciting one and the same sample site (S-on-1) for 50 fs pulses at two different pressures. The sample was a single HfO_2 layer deposited on super-polished fused silica substrate.

The low pressure data in Fig. 2 were taken after reaching base pressure. This pressure was achieved after 18 hours of pumping, during which time $F(\infty)$ dropped asymptotically to the observed value. The chamber was not heated, so the background gas was primarily water vapor (see Fig. 1b) that slowly desorbs from the interior walls of the vacuum chamber. The influence of individual gases on $F(\infty)$ was determined by re-introducing them after reaching base pressure. The results are shown in Fig. 3. Nitrogen and toluene do not affect the breakdown threshold $F(\infty)$. The outcome with toluene is very different from the observation with ns pulses, where organic compounds formed graphitic deposits that accelerated failure [5,7].

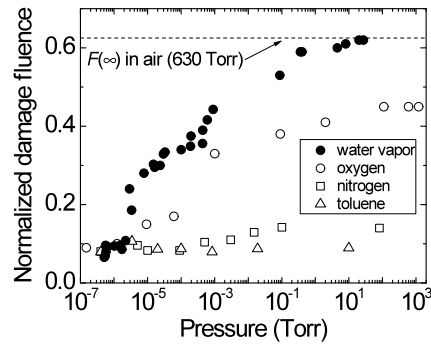


Fig. 3. Multiple pulse damage fluence of hafnia films as a function of various gas pressures. The fluences are normalized to $F(1)$

Figure 3 shows that $F(\infty)$ is sensitive to both the pressure of water vapor and gaseous oxygen. The increase in $F(\infty)$ when these gases are introduced occurs within 5 minutes (the time required to make a measurement). No further changes in $F(\infty)$ was observed up to 5 hours (data not shown). When water vapor is added, $F(\infty)$ is constant until at a partial pressure of about $\sim 3 \times 10^{-6}$ Torr there is a step-like increase. Above this pressure, the damage threshold recovers continuously and reaches its atmospheric-pressure value at a few Torr. For comparison, a typical partial pressure of water vapor in our lab is about 4 Torr (20% humidity at 300 K [19]). Finally, the oxygen pressure also affects $F(\infty)$, but the damage fluence does not reach its atmospheric value even at 2 atm of pure oxygen. Roughly speaking, the damage fluences at different pressures of air exhibit the combined effect of oxygen and water vapor.

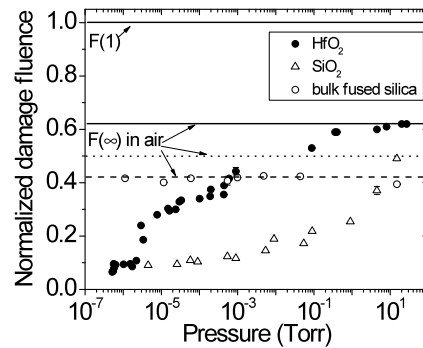


Fig. 4. Multiple pulse damage fluence as a function of water vapor pressure for three different materials: HfO_2 films, SiO_2 films and bulk fused silica. The fluences are normalized to $F(1)$ of the respective material. The horizontal $F(\infty)$ lines represent breakdown fluences under atmospheric conditions for hafnia films (solid line), silica films (dotted line) and fused silica surfaces (dashed line).

Figure 4 compares measured $F(\infty)$ values as a function of water vapor pressure of hafnia and silica films, prepared by DIBS, as well as fused silica (surface). The breakdown fluence of fused silica surfaces does not depend on ambient pressure. While $F(\infty)$ of silica films also drops with the water vapor pressure, the step-like change at low pressure observed for hafnia is absent and most of the changes occurs at higher pressures compared to hafnia films. It should be noted that the damage fluence as a function of pressure effects observed with DIBS hafnia films were also observed with hafnia films prepared by atomic layer deposition (ALD).

4. Discussion

The single fs pulse damage fluences of high-quality films and dielectric materials in general are determined by fundamental material parameters, such as multi-photon and impact ionization coefficients [20,21]. As a result, the thresholds are very deterministic and controlled by the local pulse fluence. The critical (1-on-1) fluence was found to scale as $F(1) \approx (a+bE_g)\tau_p^k$ with band gap E_g and pulse duration τ_p for dielectric oxide films [22]. Since the material parameters are not likely to be affected by the gases used in our experiments, the damage fluence is independent of gas type and pressure. In contrast, multiple pulse damage thresholds are controlled by native and laser induced defects. Our results suggest that additional absorption sites develop at reduced water vapor and oxygen pressure under laser irradiation. Oxygen and water vapor diffusing out of the film at lower pressure without laser irradiation can also change the effective multi-photon and impact ionization coefficient. However, such changes were too small to be detected with our experimental uncertainty of 3% for $F(1)$ measurements.

Figure 5 compares damage sites at atmospheric pressure and vacuum (3×10^{-7} Torr) observed with hafnia films. In atmospheric environment, damage is initiated in the beam center where the intensity is at the maximum, supporting the notion of a deterministic process. At low pressure, damage initiates at random sites within the excited sample area, not related to the region of maximum intensity. This suggests that absorption sites form at random film locations. Randomly located damage initiation sites were not observed with silica films at low water vapor pressure.

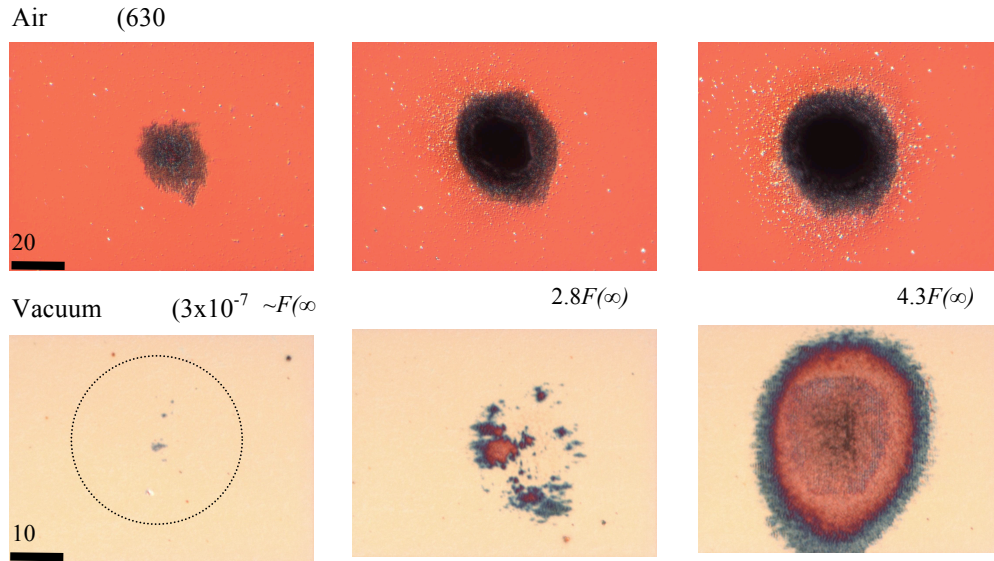


Fig. 5. Images of damaged sample sites in atmospheric (top row) and vacuum (bottom row, 3×10^{-7} Torr) environments taken with an optical microscope operating in Nomarski mode. $S = 120,000$ pulses for each exposure and the fluence is given in terms of $F(\infty)$. The sample was a single HfO_2 layer deposited on bulk fused silica

If the ambient (air) pressure is decreased oxygen can diffuse out of the film producing defects of certain concentration N_{ox} based on oxygen deficiency. One possible example is a vacancy defect that is known to exist in hafnia [23,24]. The steady state density of these defects reached at a certain pressure depends on the oxygen partial pressure p_o and the partial pressure of water vapor p_w . The dependence of N_{ox} on p_o is obvious if oxygen diffusion is driven by a concentration gradient. Water in the film and at the surface can act as a barrier for the oxygen diffusion and moreover can replenish oxygen in the film when chemisorbed. These processes are mediated by laser radiation (a pulse train of 1 kHz repetition rate). The

resulting increase of N_{ox} with decreasing pressure leads to a decrease in the multiple pulse damage threshold according to our model of laser-induced dielectric breakdown [16]. Damage is still deterministic with respect to the input fluence, that is, it starts at the beam center. The damage morphology is illustrated in Fig. 5 (top row).

Finally, it has been found experimentally that optical mirrors based on $\text{HfO}_2/\text{SiO}_2$ multilayers used in the Jefferson Lab free electron laser (FEL) exhibit lower absorption losses when the surface layer (exposed to vacuum) is HfO_2 rather than SiO_2 [25]. This is consistent with the observation in Fig. 4 that the decrease in $F(\infty)$ occurs at higher pressures in SiO_2 than in HfO_2 , suggesting that diffusion rates of molecular oxygen-related defects in SiO_2 are larger.

These deterministic processes of laser-driven defect accumulation can explain the observed continuous drop of $F(\infty)$ in HfO_2 with decreasing pressure only down to the critical water vapor pressure, $p_w = p_c \sim 3 \times 10^{-6}$ Torr, where a step-like drop in $F(\infty)$ was observed, cf. Fig. 3. At this critical pressure, the damage morphology changes, and the initiation sites occur randomly within the beam area, as was observed, cf. Fig. 5 (bottom row). Several processes can happen. Under laser radiation (1 kHz) water may not be able to form an epilayer on the surface and the surface can become charged locally. Charging of dielectric surfaces under nanosecond pulses irradiation [26-28] was reported previously. The local charges (maybe together with defects from oxygen deficiencies) can produce absorbing surface states that act as damage initiation sites. This must happen preferentially at randomly distributed sample sites. These predisposed sites could for example be boundaries between different material phases (crystalline and amorphous) or between micro-crystallites. On the other hand, it is known that silica is less prone to developing partially crystalline domains compared to hafnia, which may explain why the damage morphology in silica did not change to a random pattern at low water vapor pressure.

5. Summary

The single fs pulse (1-on-1) damage fluence $F(1)$ of dielectric oxide films (hafnia and silica) is not affected by the ambient gas pressure. The multiple pulse threshold fluence $F(\infty)$ for hafnia (silica films) decreases relative to $F(1)$ with decreasing atmospheric pressure to about 10% of $F(1)$ at 10^{-7} Torr compared to ~65% (~50%) at 630 Torr (atmospheric pressure). The decrease of $F(\infty)$ with decreasing air pressure correlates with the water vapor and oxygen content of the ambient gas with the former having the larger effect. The decrease in $F(\infty)$ is likely associated with an accumulation of defects derived from oxygen deficiency, for example vacancies. From atmospheric air pressure to pressures of $\sim 3 \times 10^{-6}$ Torr, the damage “crater” starts deterministically at the center of the beam and grows in diameter as the fluence increases. At pressure below 3×10^{-6} Torr, damage is initiated at random “sites” within the exposed area in hafnia films, while the damage morphology remains deterministic in silica films. These sites are likely created at predisposed sample locations (for example boundaries between different material phases) as a result of charging the film’s surface under laser radiation. This produces absorbing states distributed randomly across the film. The change in damage morphology was not observed with silica films, which are known to exhibit a greater degree of amorphicity than hafnia films. The gas and pressure effects are not observed with bulk fused silica surfaces.

In applications where multiple pulse damage thresholds of dielectric coatings under low pressure are a concern adding a small amount of water vapor ($\sim 10^{-3}$ Torr) if permitted by the experimental conditions can increase the damage threshold by a factor about 3.

Acknowledgments

The authors gratefully acknowledge funding by ONR, award No. N00014-06-1-0664 and No. N00014-07-1-1068, and NSF, award No. PHYS-0722622. We are grateful to Dr. Detlev Ristau (Laser Zentrum Hannover) for providing us with comparison hafnia (IBS) samples and to Dr. Joseph J. Talghader (University of Minnesota) for hafnia (ALD) samples. We thank Dr. Mark Mero for many helpful discussions.